Application of Bonded Multilayer Technology to Relaxor-Based Single Crystals for Imaging Transducer Applications

Final Technical Report

November 26, 2001

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14. ABSTRACT

The objective of this program was to evaluate the performance of single crystal materials processed in the form of multilayer crystals and crystal-composites. These multilayers have been fabricated using a process originally developed for polycrystalline ceramic. All of the multilayers were found to function with the expected four- and nine-fold increase in the effective dielectric constant. The coupling was found to vary considerably between different multilayer sizes and between different array element samples diced from the same multilayers. SEM analysis of the multilayers and of bulk material found evidence of crystal defects which likely contributed to some degradation in performance. The crystal- and crystal-composite multilayers were found to be considerably better than similar multilayers fabricated with ceramic materials. The KLM model has been used to predict the performance of array elements. Using the measured properties for 2-layer crystals and 3-layer crystal-composites, very wide bandwidth (>120%) and very good insertion loss (~18 dB) have been modeled. Because of the lower coupling measured for the crystal-composites, they did not show better bandwidth or sensitivity than the multialyer crystal. Improvements in crystal quality and a corresponding decrease in processing damage should improve the performance of multilayer crystal-composites, resulting in even better array performance.

15. SUBJECT TERMS

Single crystal relaxor ferroelectric materials; piezoelectric materials; multilayer; composites; array elements; imaging arrays

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SUMMARY

The objective of this program was to evaluate the performance of single crystal materials processed in the form of multilayer crystals and crystal-composites. Applications initially targeted included arrays for medical and underwater imaging. Early in the program it was determined that medical imaging arrays provided the best prospects for early commercialization. Thus, all efforts were directed at evaluating multilayers which could be used in transducers for medical applications.

All of the Phase I technical objectives for this program have been met. Crystal and crystal-composite multilayers have been fabricated using a process originally developed for polycrystalline ceramic. All of the multilayers were found to function with the expected four- and nine-fold increase in the effective dielectric constant which results from two- and three-layer structures, respectively. The coupling was found to vary considerably between different multilayer sizes and between different array element samples diced from the same multilayers. SEM analysis of the multilayers and of bulk material found evidence of crystal defects which contributed to some degradation in multilayer performance. The performance of crystal and crystal-composite multilayers was found to be considerably better than what has been achieved for similar multilayers fabricated with ceramic materials.

The KLM model has been used to predict the performance of array elements which utilize the multilayer structure. Using the measured properties for 2-layer crystals and 3-layer crystal-composites, very wide bandwidth and very good insertion loss have been modeled. The benefit of the crystal-composite over the crystal is not immediately obvious, primarily due to the lower coupling found for the composites.

DESCRIPTION OF RESULTS

Characterization of Single Crystal Plates

TRS Ceramics and H.C. Materials both supplied PMN-PT plates for this program. The plates and array elements cut from them were characterized to determine the properties and variation for each material. Multilayers were fabricated exclusively from the H.C. Material because the TRS material was received later in the program.

Table 1 shows the average measured properties and standard deviation for plates from the two crystal growers. From H.C. Materials, 98 plates were measured. From TRS Ceramics, 26 plates were measured. The TRS crystal has a slightly higher free and clamped dielectric constant, but it also has a higher standard deviation. The coupling, impedance, and velocity are very close. The slightly lower density of the TRS crystal may be an anomaly due to lower thickness uniformity on those plates. Further measurements are necessary to confirm this.

Characterization of Single Layer k_{33}' Array Elements

Array elements were cut from some of the plates (Table 2). The width of the elements was made to be half the plate thickness, i.e. the aspect ratio of the elements was 0.5 in all cases. The free dielectric constant of the TRS crystal was considerably higher than the H.C. crystal, as was its standard deviation. For the clamped dielectric, the H.C. crystal was slightly higher due to its lower coupling. The average coupling of the TRS material was considerably higher and the

Table 1) Average measured plate mode properties of PMN-PT plates from H.C. Materials (98 plates) and TRS Ceramics (26 plates).

	ε_{33}^T	ε_{33}^S	k_t	Z (MRayl)	v (mm/μs)	ρ (g/cc)
TRS	6607	910	0.57	37.0	4.72	7.83
std. dev	1002	106	0.01	0.2	0.05	0.06
HC	5610	862	0.57	37.2	4.67	7.97
std. dev	841	35	0.02	0.2	0.06	0.10

Table 2) Average measured array element mode properties for PMN-PT from H.C. Materials and TRS Ceramics.

	ε_{33}^T	ε_{33}^S	k_{33}'	Z (MRayl)	v (mm/μs)
TRS	5989	722	0.89	30.2	3.79
std. dev	1243	143	0.03	3.4	0.28
HC	4006	775	0.85	28	3.45
std. dev	657	130	0.05	1.6	0.14

standard deviation considerably lower than the H.C. crystal. In array element mode, the velocity and impedance of the H.C. crystal was lower.

Array elements were studied with the aid of an SEM to look for reasons for the observed variation and the lower coupling of the H.C. crystal. Table 3 shows the properties measured for the actual plates examined with the SEM. Figure 1 shows two samples from TRS. Neither of the samples is free of defects. Element 1C has a lower dielectric constants, coupling, and velocity than does element 6E. It appears that the defects of element 1C, which propagate for a larger distance, are more detrimental to the performance of the resonator. Figure 2 shows two samples from H.C. Materials. Array element C5 shows very large defects. The upper left region shows extensive cracking, and there are multiple regions of pullout. It is thought that these were caused by dicing which exposed regions of instability in the crystal, e.g. regions where the crystal growth was incomplete or where cracks had already formed. Element C9 shows smaller defects, similar to what was seen in TRS element 6E, but fewer of them. Measurements showed similar dielectric constants and velocity for C5 and C9, but the coupling for C5 is significantly lower. Thus it appears that the large defects are primarily affecting the coupling.

Characterization of Multilayer k_{33}' Array Elements

Plates of the H.C. PMN-PT were prepared to make 2-layer and 3-layer crystals and crystal-composites from plates of varying thickness (0.1, 0.15, 0.2, and 0.3 mm). Composites were designed so that the aspect ratio of the crystal strips was constant for each plate thickness. Table

Table 3) Measured properties of array elements shown in SEM photos of figures 1 and 2.

	ε_{33}^T	ε_{33}^S	k_t	Z (MRayl)	v (mm/μs)
TRS #1C	5838	698	0.90	28.2	3.47
TRS #6E	6685	741	0.92	28.9	3.51
H.C. #C5	5182	693	0.73	29.1	3.54
H.C. #C9	5169	646	0.92	28.5	3.53

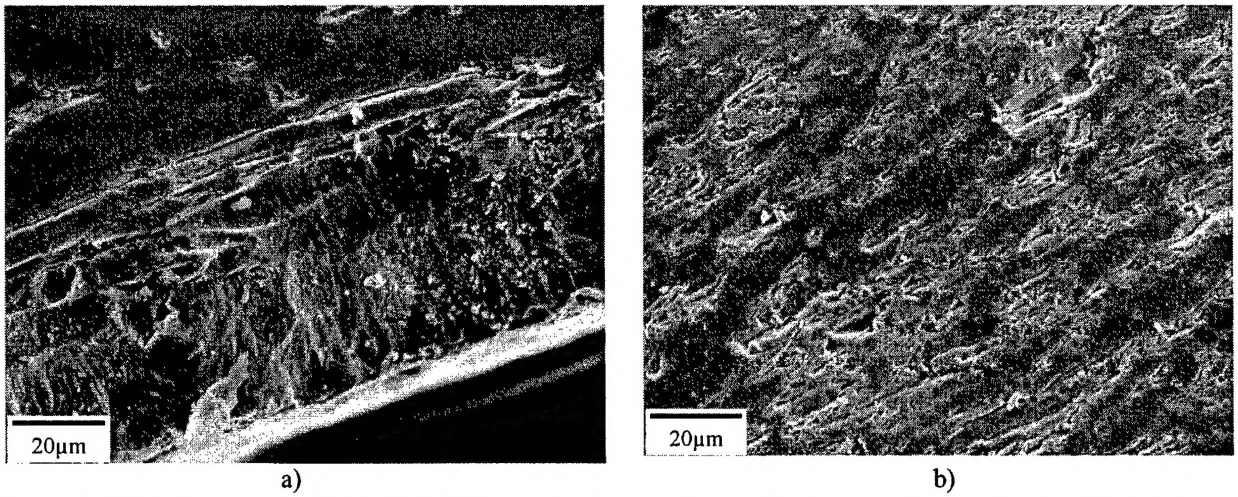


Figure 1) SEM micrographs of TRS PMN-PT crystals: a) array element 1C; b) array element 6E. See Table 3 for measured properties.

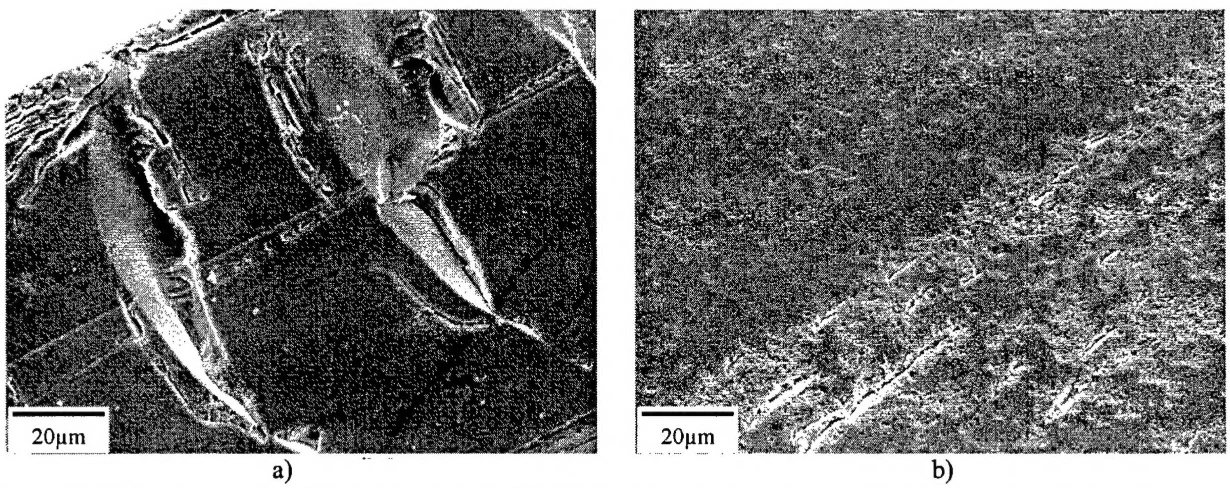


Figure 2) SEM micrographs of H.C. Materials PMN-PT crystals: a) array element C5; b) array element C9. See Table 3 for measured properties.

4 shows the dimensions to which the composites were diced for each plate thickness.

Five plates of each of four thickness were diced into composites, and five other plates of each thickness were lapped and polished for multilayer crystals. Because of the limited number of plates, it was not practical to prepare plates using our automated lapping and polishing machine. All of the plates were prepared by hand lapping and polishing. Electrodes in the form of Chrome-Gold were sputter deposited on each piece. The goal was to build one 2-layer and one 3-layer from each plate thickness of both composite and crystal. Because of the difficulty of preparing the samples, particularly for the thinnest plates, not all combinations were successfully fabricated.

Table 5 shows the measured data for crystal and crystal-composite k_{33}' resonators. The thickness denoted in the tables is the total thickness of the multilayer. With the exception of the 0.4 mm thick 2-layer crystal, all of the multilayers showed roughly the expected four- or nine-

Table 4) Dimensions of 2- and 3-layer crystals and crystal-composites

1-layer plate tk	(mm)	0.10	0.15	0.20	0.30
element width	(mm)	0.10	0.08	0.10	0.15
kerf	(mm)	0.05	0.23	0.30	0.45
pitch	(mm)	0.15	0.30	0.35	0.45
2-layer tk	(mm)	0.20	0.30	0.40	0.60
2-layer AR		0.50	0.50	0.50	0.50
3-layer tk	(mm)	0.30	0.45	0.60	0.90
3-layer AR		0.33	0.33	0.33	0.33

Table 5) Measurements of crystal and crystal-composite multilayers

Sample	Thickness	ε_{33}^{T}	ε_{33}^{S}	k_{33}'	Z (MRayl)	V (mm/μs)
			crystal multilayers			
2-layer	0.2mm	12511	3341	0.75	26.3	3.17
	0.3mm	15897	3406	0.81	27.4	3.22
	0.4mm	7365	1627	0.75	26.2	3.38
	0.6mm	15521	3195	0.83	28.3	3.31
	0.95mm	22201	3379	0.87	26.7	3.22
3-layer	0.6mm	38094	8099	0.79	23.1	2.85
			composite multilayers			
2-layer	0.3mm	8538	3478	0.70	16.7	2.78
3-layer	0.45mm	18957	7020	0.72	17.1	2.83
	0.6mm	21822	5704	0.76	17.1	2.77
	0.9mm	23808	5634	0.75	16.7	2.78

fold increase in the effective dielectric constant. The coupling ranged between 70% and 87%. In general, the thicker plates had higher coupling. As designed, the acoustic impedance of the crystal-composites was considerably lower than that of the crystals. This was due to not only a lower density, but also a lower velocity for the composites.

For comparison with the single crystal data, Table 6 shows the data generated under a separate program for ceramic and ceramic-composite multilayers. The nominal dielectric constants are roughly equivalent between the crystal and ceramic materials. The lowest coupling measured for the crystals is about the same as the highest coupling measured for the ceramics. The highest coupling of the crystals is about 23% higher than the highest coupling of the ceramic. Because of the lower velocity for the crystals, the average acoustic impedance is about 10% lower for the crystals. This should improve acoustic matching and result in arrays with wider bandwidth and higher sensitivity. Overall the performance improvement seen with crystals is considerable and should lead to arrays with improved bandwidth and sensitivity relative to what can be achieved with conventional piezoceramic materials.

SEM Analysis of Crystal Multilayers

Individual multilayers were selected for evaluation with an SEM. None of the elements showed any sign of disbond or thick bond layer. Thus the problems with plating that hindered early multilayers have been addressed, and the fabrication process which was developed for polycrystalline ceramic works equally well for single crystals. What was found through

Table 6) Measurements of ceramic and ceramic-composite multilayers (ref. TRP Core Tech Program)

Sample	Type	ε_{r33}^{S}	k_{33}'	Z (MRayl)	V (mm/μs)
1-layer	ceramic	1263	0.71	31.6	4.05
2-layer	ceramic	4860	0.7	28.5	3.65
3-layer	ceramic	8732	0.62	27.5	3.53
1-layer	composite	871	0.69	19.5	3.67
2-layer	composite	3835	0.66	17.9	3.45
3-layer	composite	9721	0.63	18.0	3.38

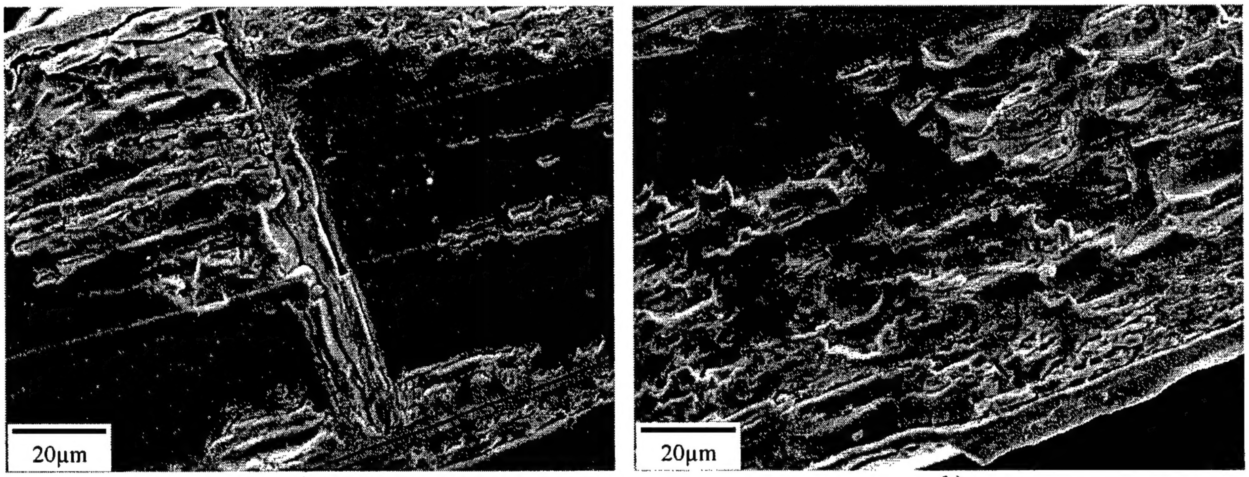
examination was the presence of large damaged regions of crystal. A description of the damage as related to the measured properties follows.

Table 7 lists the measured properties for the specific samples which were examined. Figure 3 shows damaged regions for two different array elements sectioned from the same double layer crystal. The damaged region of Piece 1 extends through the thickness of one of the layers, while the damaged region of Piece 2 is more towards the surface. The coupling and clamped dielectric constant of Piece 1 is significantly lower, indicating that the type of damage affects the amount of property degradation. It is believed that the damage has exposed regions of crystal where the growth was incomplete or where micro-fractures have formed during the growth process. Tetrad has extensive experience dicing single crystals. The conclusion drawn from that experience is that high quality crystal will show little or no damaged regions when processed carefully, but that lesser-quality crystal will expose defects when subjected to the stress of the sectioning process.

Figure 4 shows SEM photos of two crystal-composite multilayers, both made with 0.15 mm thick plates. The light region is the crystal. Figure 4a is a 2-layer, while Figure 4b is a 3-layer crystal-composite. The coupling, impedance and velocity are close for both samples, and the dielectric constant is as expected. From the SEM it is apparent that the 3-layer has a misalignment of the crystal regions of about 15%. This is near the point where bending modes are introduced which can degrade the coupling, but it does not appear to affect the coupling significantly. It is also apparent from both photos that there are regions of damage in both samples, particularly towards the outside of the crystals. It appears that on the individual plates the damage is more extensive on the 2-layer. However, since the three-layer has one more plate there is a roughly equivalent percentage of damage on both samples. It is also interesting to note that there is also damage along the vertical edges of the crystal (next to the interface with the polymer region). Thus any type of processing appears to degrade the crystal, including dicing of the composite kerfs, lapping and polishing of the surface, and sectioning of the array elements.

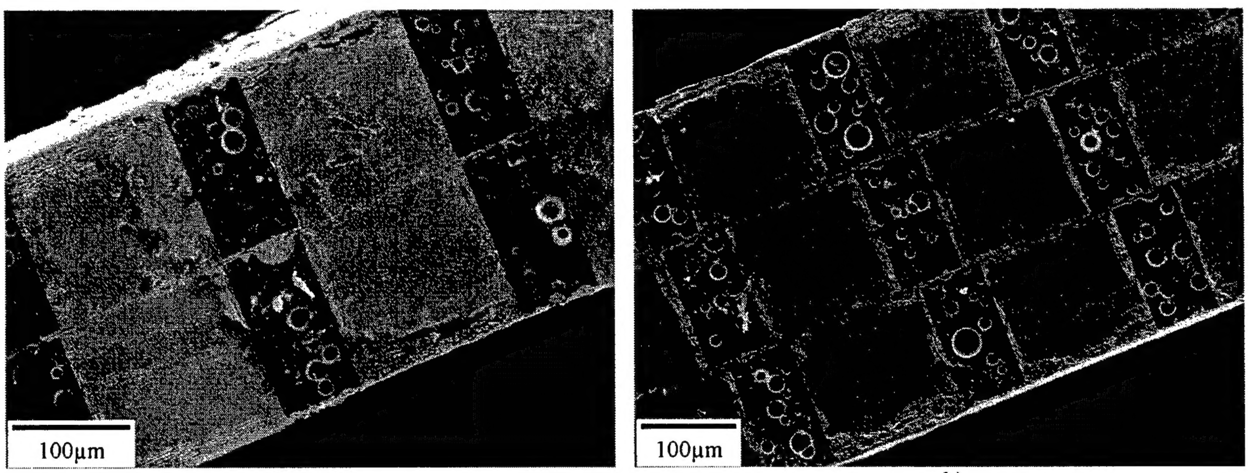
Table 7) Measured properties of array elements shown in SEM photos of Figures 3 and 4

	ε_{r33}^{T}	ε_{r33}^{S}	k_t	Z (MRayl)	V (mm/μs)
CrystDL4p1	10670	3070	0.71	26.1	3.07
CrystDL4p3	11842	3609	0.78	25.6	3.17
CompDL8	8538	3478	0.70	16.7	2.78
CompTL6	18945	6738	0.72	16.7	2.79



a) b)

Figure 3) SEM micrographs of H.C. PMN-PT 2-layer crystal multilayers, 0.1 mm plates (CrsytDL4): a) piece 1; b) piece 3. See Table 7 for measured properties.



a) b)

Figure 4) SEM micrographs of H.C. PMN-PT 2-layer and 3-layer crystal-composite multilayers: a) CompDL8, 0.15 mm plates; b) CompTL6, 0.15 mm plates. See Table 7 for measured properties.

Analysis of Transducer Performance Improvement Calculated from Measured Crystal Multilayer Properties

The KLM model has been used with the measured properties for multilayer crystals and crystal-composites to evaluate the performance improvement which is attainable. Figure 5 shows a simulation based on the highest coupling measured for any of the multilayers, which was for a 2-layer crystal with a total thickness of 0.95 mm. The combination of high coupling and high dielectric constant leads to a bandwidth which approaches 130% with a low insertion loss. Figure 6 shows a simulation using properties which are average for the multilayer crystal-composites. The measured coupling for the composites (75%) was considerably lower than the best crystals (87%). The 3-layer crystal-composite simulation shows a similar level of performance as the 2-layer crystal. Thus it appears that the benefits of the 3-layer composite, e.g. lower acoustic impedance and higher dielectric constant, are offset by the lower coupling which appears to be caused by poor quality crystal, at least in the regions where the damage due

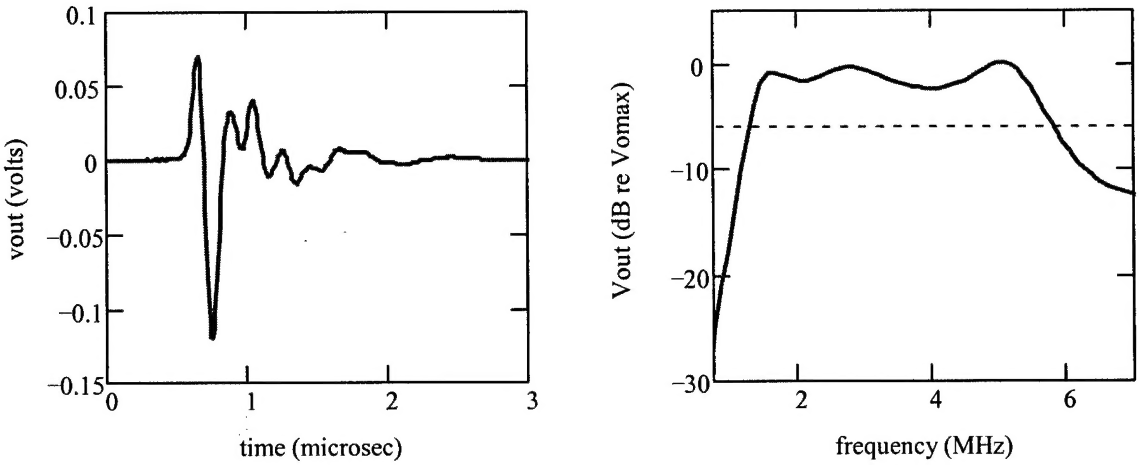


Figure 5) KLM model simulation of array response using the properties measured for a 2-layer crystal (k = 0.87, Z = 26.7 MRayl, ε_{33}^{S} = 3379, v = 3.22 mm/μs). Simulation shows BW = 128%, IL = 17.3 dB.

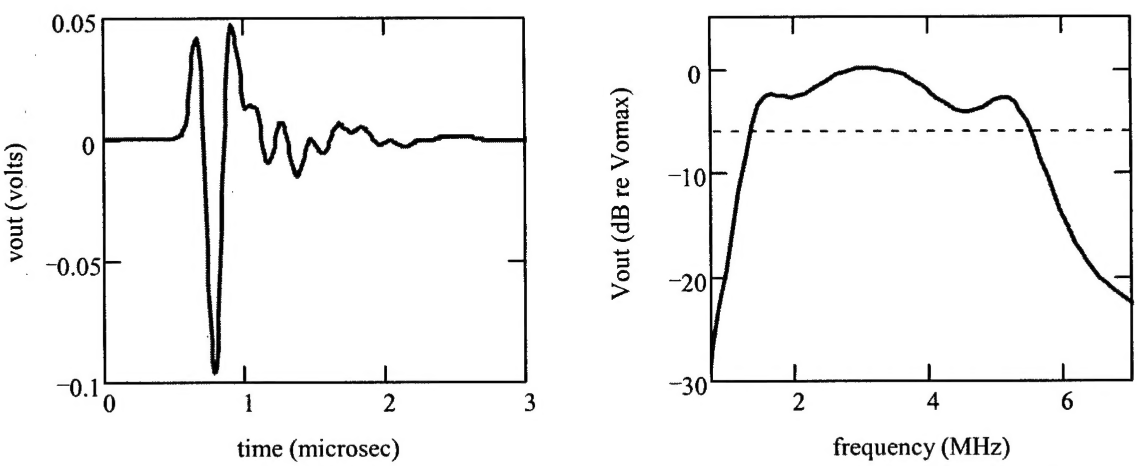


Figure 6) KLM model simulation of array response using the properties measured for a 3-layer crystal-composite (k = 0.75, Z = 17.1 MRayl, ε_{33}^{S} = 6000, v = 2.79 mm/μs). Simulation shows BW = 125%, IL = 18.8 dB.

to dicing occurred. It is thought that higher coupling should be possible if damage can be minimized or eliminated. This should result in improved performance, i.e. higher bandwidth and sensitivity.

CONCLUSIONS

A large number of single crystal plates, delivered from two sources, have been measured and the variations of properties from plate to plate and of array elements from various plates observed. It was found that for plates the PMN-PT from H.C. Materials had a slightly lower dielectric constant than similar material from TRS Ceramics, but it also had less variation amongst the plates. For array element resonators the TRS crystal again had a higher dielectric but with more variation. However, the coupling of the TRS was also considerably higher with less variation. SEM analysis has shown that both materials suffer from regions where the crystal quality appears to be poor, as indicated by regions where cracks or voids exist or other damage is

induced from lapping or polishing. In general, the quality of the H.C. crystal appears to be poorer than the TRS crystal. It appears that crystal quality becomes increasingly important as the scale of the resonators becomes smaller. It is believed that this is why the bulk plates and lower frequency multilayers performed better.

A series of crystal and crystal-composite multilayers have been fabricated using a process developed for polycrystalline ceramic materials. Initial problems with plating adhesion to the polished surfaces have been addressed. SEM analysis found no evidence of thick bond layers or delamination of the individual crystal layers. All of the samples had the expected four- or nine-fold increase in the effective dielectric constant. Thus the multilayer fabrication process is well adapted to crystals. In general, the lower frequency multilayers performed better. Again, evidence was found of dicing damage likely caused by poor crystal quality. It is thought that this damage is responsible for the lower coupling measured for thinner crystal and all of the crystal-composite multilayers.

Simulations based on the measured properties show that relative to ceramics and single layer crystals, the multilayer structure is capable of delivering very high bandwidth with very low insertion loss. Unfortunately the benefits of the multilayer crystal-composites over that of multilayer crystals are not immediately obvious, primarily because of the reduced coupling measured for these samples. It is thought that better quality crystal should be capable of being formed into the fine dimensions necessary for operation of multilayer composites with high coupling (i.e. > 82%) in the frequency range used for medical imaging arrays.

FUTURE PLANS

The technology delivered under this program has been found to be useful for developing products, particularly arrays for medical imaging applications. Based on simulations, these arrays have been shown to have good potential for improved performance relative to what can be achieved with conventional ceramic materials applied to the same multilayer configuration. It is hopeful that, once the crystal quality issues have been addressed by crystal growers, these multilayer configurations will be found in commercial products.

ITEMIZED MAN HOURS AND COSTS

Labor	$16,894.70
Labor Overhead	$19,523.51 (115.56% *)
Total Labor and Overhead	$36,323.61
Materials	$25,347.57
Equipment	$ 0.00
Consultants	$ 192.50
Subcontracts	$ 0.00
Other	$ 1,888.33
Total Other Direct Costs	$27,428.40
Total Costs Before G&A	$63,752.01
General and Administration (G&A)	$37,288.55 (58.49% *)
Total Cost	$101,040.56
Total Amount of Contract	$ 97,303.00
Difference	$ -3,737.56

* Note: Labor Overhead and G&A Rates reflect actual Tetrad rates as of October 31, 2001.

CONTRACT DELEVERIES STATUS

This final report, with form SF298 and DD250 attached, completes the requirements for this contract.

REPORT PREPARER

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DECLARATION OF TECHNICAL DATA CONFORMITY

The Contractor, Tetrad Corporation, hereby declares that, to the best of its knowledge and belief, the technical data delivered herewithin under contract No. DAAH01-01-C-R141 is complete, accurate, and complies with all requirements of the contract.

Date: 11-26-2001

Michael J. Zipparo, Ph.D.
Research and Development Engineer

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